

Standardization of Carbon-14 by $4\pi\beta$ Liquid Scintillation Efficiency Tracing with Hydrogen-3

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Carbon-14 in the form of [¹⁴C]tartaric acid solution has been standardized by means of $4\pi\beta$ liquid-scintillation efficiency tracing using the NBS [³H]water standard. The method of computing the detector efficiency for a two-phototube counting system using a standard of ³H is described. The combined uncertainty in the ¹⁴C radioactivity concentration, which may be treated as if it were one standard deviation, is 0.20%.

1. Introduction

The method of standardizing one β -particle-emitting nuclide using a standard of another to establish the counting-system efficiency is evolving from work in several laboratories.⁽¹⁻⁶⁾ A review of this method for a single-phototube system as well as a discussion of other more-established coincidence techniques is given in the latest edition of the National Council on Radiation Protection and Measurements (NCRP) Report 58, 1985 edition.⁽⁷⁾

Liquid-scintillation counters have long been used for comparative measurements of low-energy β -emitters because of their high efficiencies; coincidence-counting efficiencies are typically in excess of 50% for [³H]water and 94% for ¹⁴C-labelled compounds in aqueous samples in commercial scintillators and two-phototube systems. The efficiency is somewhat higher if one removes the coincidence requirement and sums the individual outputs from the two tubes. But, this gain in efficiency is at the expense of an increase in the numbers of detected "after pulses" from the individual phototubes,⁽⁸⁾ which are almost entirely removed by the coincidence requirement. Various methods of extrapolation of the integral counting rate vs discrimination level have been proposed,⁽⁹⁻¹¹⁾ but as Houtermans has pointed out "any extrapolation method is just as good as the physical model, justifying the adopted shape of the extrapolated function".⁽²⁾ The method of β -particle efficiency tracing is based on comparing the activity

of an unknown with that of a standard, in aliquots of the same liquid scintillator and in the same system. The standard is used to calibrate the system and is linked to the detection efficiency for the unknown by computing the detection efficiencies, using theoretical β -ray spectral distributions, for both the standard and the unknown. Here [³H]water is the standard and [¹⁴C]tartaric acid the unknown. A computed ionization-quenching correction is made using the theory of Birks,⁽¹²⁾ and from this and the computed efficiency the figure of merit (to be defined below) can be calculated. The ¹⁴C sample is now measured under similar geometrical and quenching conditions and the previously measured figure of merit from the standard allows its total activity to be computed. This, in rather broad strokes, illustrates the principle of the method of liquid-scintillation efficiency tracing. It is necessary, however, to consider the detailed processes involved in more detail.

2. Description of the Method

If electrons expend energy in the scintillating solution of a liquid-scintillation counting system, then they will be detected if photons, with energies in the near u.v., are incident on the photocathode in sufficient numbers that one or more photoelectrons strike the first dynode of the phototube. But only a very small fraction of the photons produced in the scintillator will be incident upon the photocathode,

and not all the photoelectrons emitted will reach the first dynode. There will thus be an average threshold energy below which electrons will generate insufficient photons to enable more than an occasional photoelectron to be incident upon the first dynode.

In the case of a β -particle-emitting nuclide in the scintillator, the β -particles with energies below threshold will, in general, not be detected, but, with increasing β -particle energy, first one, then two and more photoelectrons will be incident upon the dynode, and the corresponding β -decay will be registered. At any given β particle energy, E , there will be an average of $\bar{n}(E)$ photoelectrons incident upon the first dynode of the phototube, with numbers x distributed around that average value. Thus for any mean value of $\bar{n}(E)$ there will be a finite probability that no photoelectrons will strike the first dynode. In this case the primary ionizing event will not be detected. This has been aptly named "zero-detection probability." It is in the nature of an efficiency, for low energies, limited to the phototube itself and is only one factor in the overall detection efficiency.

As the β -particle energy spectrum is collected by the detector system, many low-energy events that deposit energy E' in the scintillator, such that $\bar{n}(E')$ is finite but small, will give rise to numbers of photoelectrons incident on the first dynode distributed around an average number equal to $\bar{n}(E')$. As each burst of photons incident upon the photocathode is an independent event, it is assumed that each $\bar{n}(E')$ is the mean of a Poisson distribution, in which the probability of any integral number x occurring is given by

$$P[x, \bar{n}(E')] = \frac{\bar{n}(E')^x \exp[-\bar{n}(E')]}{x!} \quad (1)$$

Thus for any value of $\bar{n}(E')$, the zero-detection probability is $P[0, \bar{n}(E')]$, so that for $\bar{n}(E')$ equal, for example, to 1, 3 and 8 the zero-detection probabilities are respectively 0.368, 0.050 and 0.0003.

Likewise, for low numbers of secondary-emission electrons from the first dynode there will be a finite probability of zero detection by the second dynode, but this represents only a second-order correction.

The next step is to derive the overall efficiency for the whole system. To do this it is necessary to invoke the so-called "figure of merit" which has been variously defined by different authors. Thus it has been defined as a factor, or its reciprocal, that relates the energy of an ionizing particle, or the energy that it deposits in the scintillator solution, and the number of photoelectrons "at" or "on" the photocathode, or "striking" the first dynode of the phototube. There will be small differences between these definitions, depending on the collection efficiency of the first dynode. In this paper the figure of merit is defined specifically as $\eta(\text{keV}^{-1})$, which is the average number of photoelectrons striking the first dynode of any one given phototube divided by the energy E of a detected electron, in kiloelectronvolts.

But, clearly a figure of merit can only be independent of energy if it is expressed in terms of the total energy of the photons produced by a given ionizing event and not in terms of the total energy of the ionizing particle, much of which can be expended outside the solution, or dissipated as heat within the solution by secondary interactions. The former loss is generally referred to as the wall effect, designated by the fraction $[1 - W(E)]$, and the latter as quenching, designated by the fraction $[1 - Q(E)]$. Thus the amount of energy *not* lost to the solution by the escape of higher-energy electrons into the walls of the container or through the upper liquid-gas interface is $EW(E)$. Of this energy deposited in the solution, an amount $EW(E)Q(E)$ is *unquenched* and is therefore totally available for the production of fluorescence photons. These photons are within a relatively narrow band of wavelength around 400 nm, and apart from variations in their point of origin in the solution, there will be little cause for variation in the figure of merit. Moreover by calibrating the system, under as identical conditions as possible, with a ^3H water standard, compensation should be very largely achieved for any such variations.

Thus if an electron of energy E deposits, in the liquid scintillator, an amount of energy of which a fraction equal to $EQ(E)W(E)$ is converted totally into photons, and if the system has a figure of merit equal to η , then the average number of photoelectrons, \bar{n} , incident upon the first dynode of the phototube will be given by

$$\bar{n}(E) = \eta EQ(E)W(E). \quad (2)$$

Therefore the counting efficiency ϵ_1 , above the detection threshold, for a single phototube system for electrons of energy E is

$$\epsilon_1 = \{1 - \exp[-\eta EQ(E)W(E)]\}, \quad (3)$$

the probability of detecting the lowest-energy events just above threshold being one minus the probability of not detecting them.

For a liquid-scintillation counting system employing two well-matched symmetrically-located phototubes in coincidence, the analogous expression for the coincidence-counting efficiency is

$$\epsilon_{1,2} = \{1 - \exp[-\eta EQ(E)W(E)]\}^2. \quad (4)$$

Quenching can be caused in several ways, such as ionization, color, and chemical. In the measurements described here, compositions of scintillator solutions were made as identical as possible so that the effects of color or chemical quenching were the same for the ^3H water and ^{14}C tartaric acid samples. The ionization quenching was calculated from the formula of Birks,⁽¹²⁾ and additional small experimental corrections made using different methods. Controlled increments of quenching were achieved by the addition of measured amounts of the aqueous sample. The value of $W(E)$ is essentially equal to unity up to energies of about 20 keV, and the effect of its decrease

thereafter on the exponential involving $\eta EQ(E)W(E)$ is overridden below 3000 keV by the increase of E .

But so far we have considered only those events above threshold that deposit enough energy in the scintillator to produce one or more photoelectrons in the phototube of a single-phototube or coincident system. In order to correct to 100% efficiency, extrapolation to zero energy of the integral or differential pulse-height distributions is often used. In the case of liquid-scintillation counting this may not be too inaccurate for high-energy β -particle emitters, because the low-energy spectral distortions caused by Poisson peaks arising from the detection of single, or two or more, electrons are relatively insignificant. But, it is precluded for low-energy β emitters by the superposition of Poisson distributions for \bar{n} equal to 1, 2, 3, . . . on what would otherwise be a smooth pulse-height distribution. This problem was, however, circumvented in the 1960's by Horrocks and Studier⁽¹⁾ and others using the Fermi probability distribution, and by Bryant *et al.*,⁽¹³⁾ using a single-photon light source with a mechanically controlled shutter to simulate the Fermi distribution.

As mentioned above, the efficiency of a given liquid-scintillation-counting system is measured using a standard, such as one of $[^3\text{H}]\text{water}$, and then the overall counting efficiency for the system (i.e. also considering the zero-detection probability) is computed using the Fermi distribution function, $P(Z, E)dE$ (the probability of emission in unit time of β -particles with energies lying between E and $E + dE$, for each energy increment dE). The overall efficiency for the scintillation-counting system operating in the coincidence mode is then given by the following equation where the integral in the numerator represents all the events between zero and maximum energy that will *probably be detected*, in unit time, and that in the denominator represents the *probable total number* of β -particles emitted in unit time in the scintillator.

$$\epsilon_c = \frac{\int_0^{E_{\beta\text{max}}} \{1 - \exp[-\eta EQ(E)W(E)]\}^2 P(Z, E) dE}{\int_0^{E_{\beta\text{max}}} P(Z, E) dE} \quad (5)$$

ϵ_c can be measured experimentally, for a given scintillator, using the ^3H standard, and the figure of merit, $\eta Q(E)W(E)$, the only unknown in equation (5), can then be computed.

Equation (5) holds for any system with a given figure of merit, and given values of $Q(E)$ and $W(E)$. Repetitive measurements will be reproducible within statistical limits *provided* that η , $Q(E)$, and $W(E)$ remain unchanged. Also, using standard liquid-scintillation vials, a series of samples may be measured with the assurance that $W(E)$ is equal to unity up to energies, E , of about 20 keV.⁽³⁾ At higher energies the negative exponential of the product $\eta EQ(E)W(E)$ is insignificantly different from

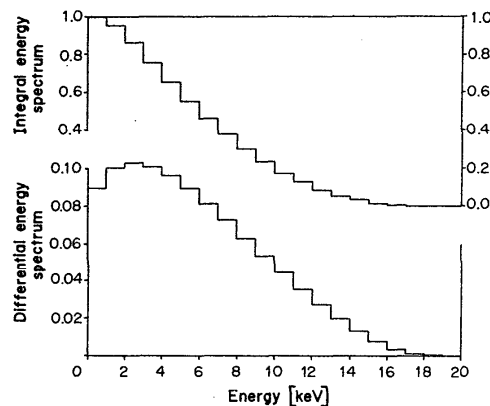


Fig. 1. Computed ^3H spectra, integral and differential, based on a Fermi distribution.

zero.⁽⁷⁾ In fact, for typical values of $Q(E)$, and values of the energy E , between 20 keV and 3.0 MeV, $[1 - \exp(-\eta EQ(E)W(E))]$ is equal to 1.0000, the increase in E greatly outweighing the decrease in $W(E)$.⁽⁷⁾

In the measurements described herein, the efficiency of a given scintillator-phototube system is calibrated using a $[^3\text{H}]\text{water}$ standard, in conjunction with equation (5), and the product $\eta Q(E)W(E)$ is evaluated as a function of the energy E , by means of a step-by-step summation in small increments, as shown in Fig. 1 for 1-keV steps. Typical values of $(\exp(-\eta EQ(E)W(E)))$ at different energies are shown in Table 1. These values of $\eta EQ(E)W(E)$, for electrons of energy E , for a given scintillator solution are independent of the electron source.

Using the same batch of scintillator solution for approximately equal quenching, ^{14}C -solution aliquots are assayed, simply from the measured count rates and the efficiencies given by equation (5) (using the ^3H -calibrated values of $\eta Q(E)W(E)$ for each energy

Table 1. Detector efficiency (column 4) for 1-keV energy bins, using η equal to 0.358 photoelectrons per kiloelectronvolt, $Q(E)$ from Ref. (19), and $W(E)$ equal to unity

Energy (keV)	$x = [\eta EQ(E)W(E)]$	e^{-x}	$[1 - e^{-x}]^2$
0.5	0.06220	0.9397	0.0036
1.5	0.26060	0.7706	0.0526
2.5	0.49168	0.6116	0.1509
3.5	0.74123	0.4765	0.2740
4.5	1.00382	0.3665	0.4013
5.5	1.27644	0.2790	0.5198
6.5	1.55727	0.2107	0.6230
7.5	1.84508	0.1580	0.7089
8.5	2.13886	0.1178	0.7783
9.5	2.43799	0.0873	0.8330
10.5	2.7596	0.0633	0.8774
11.5	3.07568	0.0462	0.9098
12.5	3.3936	0.0336	0.9340
13.5	3.7134	0.0244	0.9518
14.5	4.034	0.0177	0.9649
15.5	4.357	0.0128	0.9745
16.5	4.681	0.0093	0.9816
17.5	5.006	0.0067	0.9867
18.5	5.332	0.0048	0.9903

E , and the Fermi function appropriate to ^{14}C . For β -emitting radionuclides with end-point energies 200 keV, or above, the value of $[1 - \exp(-\eta EQ(E)W(E))]$ is so insignificantly different from unity that the coincidence-counting efficiency also reduces to unity, and the method becomes a straightforward relative measurement.

Because of the numerous factors to be considered in the application of this method, previous estimates of the uncertainties in the method have been rather conservative.^(4,6) Grau Malonda and Garcia-Toraño assigned an overall uncertainty of $\pm 3\%$.⁽⁴⁾ Coursey *et al.*⁽⁶⁾ described the standardization of ^{99}Tc using standards of three different nuclides: ^3H , ^{14}C and ^{60}Co , the results for which agreed to within 0.3%. Nevertheless, they assigned an overall uncertainty to the ^{99}Tc activity of 1.6%. This was the linear sum of the random uncertainty of the mean at the 99% confidence level, and the linear sum of estimated systematic uncertainties. It now appears that the uncertainty in the method may be, in fact, considerably lower than previously suggested.

3. Results and Discussion

Equation (5) gives the liquid-scintillation coincidence-counting efficiency for ^3H or ^{14}C as functions of the figure of merit. In the computations the integral is replaced with a summation. The precision depends somewhat on the width of the energy bins, but the method can be illustrated graphically using 1-keV bins. Figure 1 shows the theoretical integral and differential β -particle-energy distributions, as step functions for ^3H .

To compute the coincidence-counting efficiency, the differential-energy distribution (Fig. 1b) is folded with the detector efficiency for the appropriate median value of E , for a given increment dE (corresponding to each increment of the integrand in the numerator of equation (5)). This detector efficiency is given by the curve in Fig. 2 for η equal to 0.358 photoelectrons per keV. The computed output spec-

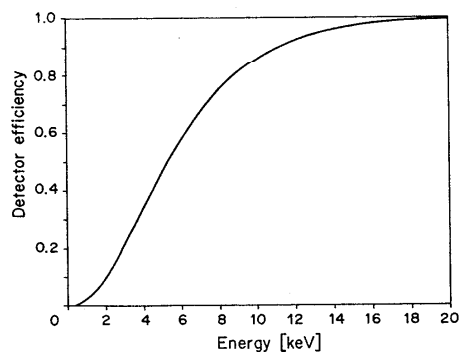


Fig. 2. Detector efficiency for a coincidence system for η equal to 0.358 photoelectrons per keV, $Q(E)$ from Ref. (19), and $W(E)$ equal to unity.

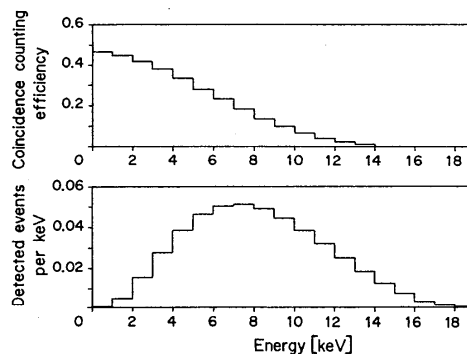


Fig. 3. The upper plot shows the counting efficiency for tritium in a coincidence counting system. The lower curve shows the differential counting efficiency per kiloelectron-volt in the same system.

tral distributions for ^3H , integral and differential, are shown in Figs 3a and b, respectively. These particular values were chosen because they lead to a computed efficiency for ^3H of 47%, which was the experimentally-observed value. It is interesting to note, from Fig. 3b, how little the events below 2 keV contribute to the counting efficiency.

The corresponding spectra for ^{14}C are shown in Fig. 4 for the region below 20 keV. The solid lines represent the theoretical spectra. It may again be noted that the increments in the number of events per keV in the low-energy region is approximately 1%. Numerical integration of this energy distribution over the detector efficiency (Fig. 2) results in the output spectra shown by dashed lines in Fig. 4. The computed coincidence-counting efficiency for ^{14}C is 93.8%. In practice, the procedures of the method take

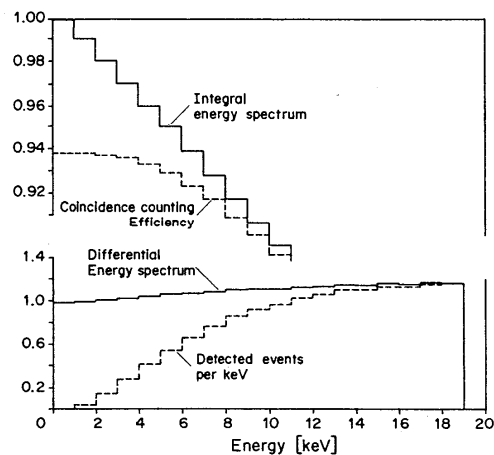


Fig. 4. Theoretical and computed output spectra for ^{14}C . The solid lines show the differential (lower) and integral (upper) plots based on a Fermi distribution. The dashed lines correspond to the differential counting efficiency per keV (lower) and integral counting efficiency (upper) for a coincidence counting system.

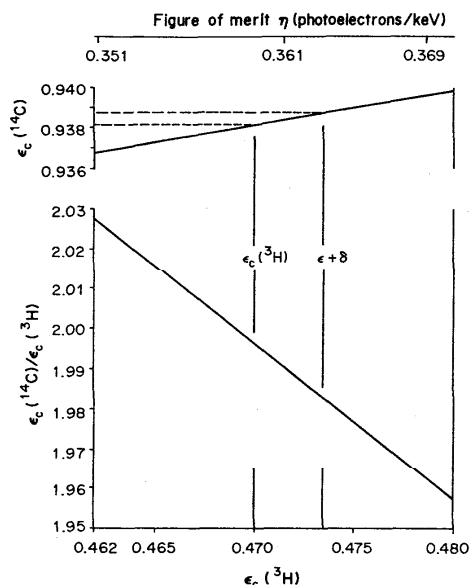


Fig. 5. Ratio of the counting efficiencies $\epsilon_c(^{14}\text{C})/\epsilon_c(^3\text{H})$ as a function of the figure of merit and the $\epsilon_c(^3\text{H})$. The vertical lines show the effect of a 0.7% uncertainty in the ^3H water standard on the computed ^{14}C counting efficiency.

on a fairly simple form. Figure 5 shows a plot of the efficiency ratio $\epsilon_c(^{14}\text{C})/\epsilon_c(^3\text{H})$ against both η , and $\epsilon_c(^3\text{H})$. Thus, with a measured efficiency for a ^3H standard, the ^{14}C efficiency can be read directly from the plot in Fig. 5.

For the present work we combined the computed efficiencies from the Atomic Energy Research Establishment—Harwell (AERE) and the Junta de Energía Nuclear—Madrid (JEN) to arrive at the following expression for the efficiency ratio vs the $\epsilon_c(^3\text{H})$:

$$\frac{\epsilon_c(^{14}\text{C})}{\epsilon_c(^3\text{H})} = a_0 + a_1\epsilon_c(^3\text{H})^{-1} + a_2\epsilon_c(^3\text{H})^{-2} + a_3\epsilon_c(^3\text{H})^{-3} + a_4\epsilon_c(^3\text{H})^{-4}, \quad (6)$$

where the a_i are determined by linear regression. Equation (6) has no physical significance, but it was useful for interpolating efficiency ratios from the

Table 3. Beta-particle efficiency tracing of ^{14}C with NBS ^3H water standard. 1983 measurements with Beckman 7800 liquid-scintillation counting system.

(a) Measurements of the NPL ^{14}C tartaric acid solution

No.	Horrock's H*	Measured ^{14}C count rate ($\text{s}^{-1} \text{mg}^{-1}$) July, 1983	Deviation from equation 7 (%)
1	68.8	22.218	0.13
2	69.5	22.161	-0.10
3	69.5	22.165	-0.08
4	69.8	22.137	0.08
5	72.0	22.137	-0.13
6	84.5	22.117	0.19
7	84.5	22.091	0.07
8	87.8	22.044	-0.03
9	93.0	21.987	-0.12

(b) Efficiencies for the NBS ^3H water standard and computed activity concentration for the NPL ^{14}C tartaric acid solution

No.	Horrock's H*	Measured $\epsilon_c(^3\text{H})$ (%)	Computed activity ^{14}C , October, 1975 (Bq mg^{-1})
1	74.0	47.88	193.68
2	74.0	48.01	193.64
3	76.7	47.62	193.61
4	77.0	47.42	193.65
5	81.0	46.39	193.75
6	81.7	46.33	193.73
Average			193.68

curve in Fig. 5. This expression fits the data to within 0.2% between $\epsilon_c(^3\text{H})$ equal to 12–86%. Solution of equation (6) for $\epsilon_c(^3\text{H}) = 47\%$ gives $\epsilon_c(^{14}\text{C}) = 93.8\%$ and it can be seen from Fig. 5 that the computed ^{14}C efficiency is rather insensitive to small changes in the ^3H efficiency. As shown in Fig. 5, a 0.7% uncertainty, δ , in $\epsilon_c(^3\text{H})$ results in only a 0.06% change in the computed ^{14}C efficiency.

Experimental data to check the validity of this model have been collected over several years with different detectors. The experimental methods and equipment will be described elsewhere.⁽¹⁴⁾ In 1983 the available data were pooled in order to measure the radioactivity concentration of the National Physical Laboratory (NPL) ^{14}C tartaric acid solution using the National Bureau of Standards (NBS) ^3H water standard.⁽¹⁵⁾ The results are given in Table 2. The good agreement between different detectors and for data taken as much as four years apart was very

Table 2. Beta-particle efficiency tracing of ^{14}C using the NBS ^3H water standard: 1979 to 1983 measurements. Activity concentration as of October, 1975

Point No.	Detector	Measured $\epsilon_c(^3\text{H})$ (%)	Computed ratio $\epsilon_c(^{14}\text{C})/\epsilon_c(^3\text{H})$	Computed $\epsilon_c(^{14}\text{C})$ (%)	Activity concentration (Bq/mg)
1	Packard 3320	30.00	2.9976	89.93	193.76
2	Packard 4530	30.31	2.9703	90.03	193.65
3	Beckman 6800	30.44	2.9591	90.08	193.31
4	Packard 4530	34.83	2.6230	91.36	193.46
5	Packard 3320	38.41	2.4009	92.22	191.48
6	Beckman 6800	40.23	2.3020	92.61	192.35
7	Packard 3320	41.25	2.2500	92.81	193.77
8	Packard 4530	43.26	2.1544	93.20	193.81
9	Beckman 6800	48.05	1.9567	94.02	191.27
10	Packard 4530	51.62	1.8320	94.57	191.59
Average					192.85

Table 4. Estimated uncertainties in the standardization of ^{14}C by β -particle efficiency tracing with NBS $[\text{H}]\text{water}$ standard

Class A ⁽¹⁷⁾	(a) liquid-scintillation counting of ^3H , $n = 6$	0.03%
	(b) liquid-scintillation counting of ^{14}C , $n = 9$	0.12%
Class B	(a) β -particle spectral distribution	0.01%
	(b) phototube mismatch	0.05%
	(c) low-level discriminator	0.10%
	(d) uncertainty in $[\text{H}]\text{water}$ standard	0.06%
	(e) uncertainty in quenching correction	0.10%
combined in quadrature		0.20%

encouraging. Following the installation of a new Beckman 7800 instrument a new set of measurements was undertaken in 1983. The data obtained for the remeasurement of the ^{14}C in 1983 are shown in Table 3. The quench correction was made using the Horrock's H number, H^* ,⁽¹⁶⁾ which is a ratio of the half height of the Compton edge for an external ^{137}Cs – ^{137m}Ba source, for the sample to an arbitrary unquenched standard. For the ^{14}C data in Table 3, the counting rate per milligram is expressed as a linear function of the H^* :

$$N_{\beta}(^{14}\text{C}) = a + b H^*, \quad (7)$$

where $a = 22.68 \text{ s}^{-1} \text{ mg}^{-1}$ and $b = 7.20 \times 10^{-3} \text{ s}^{-1} \text{ mg}^{-1}$. Tritium measurements for six samples are shown in Table 3b. Counting rates calculated for ^{14}C using equation (7) may be compared directly with the six $[\text{H}]\text{water}$ samples at the same H^* .

The agreement between the 1982 value of $192.85 \text{ Bq mg}^{-1}$ for the $[\text{H}]\text{water}$ concentration and the 1983 value of $193.68 \text{ Bq mg}^{-1}$ is quite satisfactory, but the latter value is to be preferred because of the better experimental conditions.

The uncertainty in the present result has been estimated according to the recommendations of the Bureau International des Poids et Mesures.⁽¹⁷⁾ Class A uncertainties are those which may be estimated from repetitive measurements. In Table 3 we have two such uncertainties; associated with the ^3H , for which the standard deviation for six samples is 0.03%, and with the ^{14}C , for which the standard deviation for nine samples is 0.12%. The Class B uncertainties (formerly called "systematic uncertainties") have been estimated as if they were standard deviations. Table 4 lists the estimates of the Class A and B uncertainties separately: combining them in quadrature gives an estimated "combined uncertainty" of 0.20%.

The choice of the functions $Q(E)$ and $W(E)$ introduces negligible uncertainty in the present result. The expressions used for $Q(E)$ at the AERE and JEN

have been published previously.^(18,19) If $Q(E)$ is taken as unity from 0 to 150 keV, it results in only a 0.1% change in the efficiency for ^{14}C as computed using equation 5. For lower energy β -particle emitters such as ^{241}Pu and ^{63}Ni , the choice of $Q(E)$ is significant. Recent measurements of $Q(E)$ have been reported by Rundt *et al.*,⁽²⁰⁾ using the NBS $[\text{H}]\text{toluene}$ standard.⁽²¹⁾ Work is also underway at the JEN and NBS to examine the influence of $Q(E)$ on computed efficiencies of ^{99m}Tc Auger electrons.

References

- Horrocks D. L. and Studier M. H. *Anal. Chem.* **33**, 615 (1961).
- Houtermans H. *Nucl. Instrum. Methods* **112**, 121 (1973).
- Gibson J. A. B. and Gale H. J. *J. Phys. E* **1**, 2, 99 (1968).
- Grau Malonda A. and Garcia-Toraño E. *Int. J. Appl. Radiat. Isot.* **33**, 249 (1982).
- Ishikawa H., Takiue M. and Aburai T. *Int. J. Appl. Radiat. Isot.* **35**, 463 (1984).
- Coursey B. M., Gibson J. A. B., Heitzman M. W. and Leak J. C. *Int. J. Appl. Radiat. Isot.* **35**, 1103 (1984).
- NCRP Report 58, *A Handbook of Radioactivity Measurements Procedures* 2nd edn, (Ed. Mann W. B.) p. 199, (NCRP Publications, Washington, D.C., 1985).
- Smith D. In *Applications of Liquid Scintillation Counting in Radionuclide Metrology*. BIPM Monographie 3. Chap. VI (Eds Mann W. B. and Taylor J. G. V.) (BIPM, Sèvres, 1980).
- Steyn J. *Proc. Phys. Soc. A* **69**, 865 (1956).
- Goldstein G. *Nucleonics* **23**, 67 (1965).
- Flynn K. F., Glendenin L. E. and Prodi V. *Organic Scintillators and Liquid Scintillation Counting* (Eds Horrocks D. L. and Peng C. T.) p. 687 (Academic Press, New York, 1971).
- Birks J. B. *The Theory and Practice of Scintillation Counting* p. 105 (Pergamon Press, Oxford, 1964).
- Bryant J., Jones D. G. and McNair A. In *Standardization of Radionuclides*, STI/PUB/139, p. 47. (IAEA, Vienna, 1967).
- Coursey B. M. To be published.
- Unterwieser M. P., Coursey B. M., Schima F. J. and Mann W. B. *Int. J. Appl. Radiat. Isot.* **31**, 611 (1980).
- Horrocks D. L. In *Liquid Scintillation: Science and Technology* (Eds Noujaim A. A., Ediss C. and Weibe I. I.) p. 185 (Academic Press, New York, 1976).
- Giacomo P. *Metrologia* **17**, 73 (1981).
- Gibson J. A. B. In *Applications of Liquid Scintillation Counting in Radionuclide Metrology*, Chap. IV (Eds Mann W. B. and Taylor J. G. V.) (BIPM, Sèvres, 1980).
- Garcia-Toraño E. and Grau Malonda A. *Comp. Phys. Commun.* **23**, 385 (1981).
- Rundt K., Kouru H. and Oikari T. In *Advances in Scintillation Counting* (Eds McQuarrie S. A., Ediss C. and Wiebe L. I.) p. 30 (Univ. of Alberta, Banff, 1983).
- Garfinkel S. B., Mann W. B., Medlock R. W. and Yura O. *Int. J. Appl. Radiat. Isot.* **16**, 27 (1965).
- Grau Malonda A., Garcia-Toraño E., Los Arcos J. M. and Coursey B. M. To be published.